



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF :  
YOSHITAKA AOKI ET AL. : EXAMINER: KUO LIANG PENG  
SERIAL NO. 10/643,978 :  
FILED: AUGUST 20, 2003 : GROUP ART UNIT: 1712  
FOR: HEAT CONDUCTIVE COMPOSITE SHEET AND PROCESS FOR PRODUCING  
SAME

DECLARATION UNDER 37 CFR 1.132

HONORABLE COMMISSIONER OF PATENTS & TRADEMARKS  
WASHINGTON, D.C. 20231

SIR:

1. I, Ikuo Sakurai, a citizen in Japan residing at Annaka-shi, Gunma-ken, Japan, hereby state and declare that:

2. In March, 1987, I graduated from Department of Applied Chemistry for Resources, Faculty of Engineering, Tokyo University of Agriculture and Technology.

3. In April, 1987, I entered the employment of Shin-Etsu Chemical Co., Ltd. From 1987 to 2000, I engaged in R & D of mainly inorganic materials. Since 2000, I have engaged in R & D of organosilicone compounds and finished organosilicone compounds, such as heat conductive silicone products.

4. I have taken over the development and improvement of the technology in the field to which the above-identified patent application relates; therefore I am well versed in the inventions claimed and disclosed in the application.

5. I understand both the Japanese and English languages.

6. The following experiments were performed by me or under my control and/or supervision:

In order to show that the pre-cured upper layer 7A of Fick (US Patent No. 4,842,911) is not capable of melting at a temperature that ranges from 40°C to 100°C, and that the melting behavior of the layer (a) of the present application is

-2-

unexpected from Fick, following Experiment 1 and Comparative Experiments 1 to 3 were conducted.

Experiment 1 (heat softening, heat conductive sheet)

A heat softening, heat conductive sheet K-2' with a thickness of 2 mm was produced from the composition K-2 (page 21, line 16 and Table 1 of the specification) as follows and was used as the layer (a) of the present invention.

The composition K-2 was prepared in the same manner as described on page 21, lines 13-16 of the specification. The composition K-2 was diluted with toluene and applied to one surface of a PET film that had been treated with a silicone-based mold release agent. The applied composition on the PET film was heated at 60°C for 10 minutes and subsequently at 120°C for 10 minutes. As a result, toluene was removed from the composition, and a 200- $\mu$ m-thick sheet was obtained on the PET film. After cooled to room temperature (23°C, this also applies below), the sheet was peeled from the PET film. Ten of the peeled sheets were layered, and each sheet was thermally pressure-bonded to its adjacent sheet(s) with a roll heated to 150°C so that air bubbles were not trapped between the layered sheets. Thus, the heat softening, heat conductive sheet K-2' with a thickness of 2 mm was produced.

Fick states that, "air-dry type of curing" and "solvent-type curing" may be used in a curing step in addition to "peroxide curing" (col. 6, lines 52-61). However, the terms "air-dry type of curing" and "solvent-type curing" are not conventional in the art of silicones. Therefore, the curing mechanisms of "air-dry type of curing" and "solvent-type curing" cannot be well understood. On the other hand, it is known that "condensation curing" and "addition curing" as well as "peroxide curing" are typical curing types of silicones. Therefore, in the following Comparative Experiments, cured products of a peroxide curing silicone rubber composition, a

-3-

condensation curing silicone rubber composition, and an addition curing silicone rubber composition were used.

#### Comparative Experiment 1 (peroxide-cured silicone rubber sheet)

A peroxide curing silicone rubber composition includes organopolysiloxane molecules and peroxide compounds. When a peroxide curing silicone rubber composition is heated, the peroxide compounds are decomposed by heat to produce free radicals. The free radicals act on the organo groups bonded to silicon atoms within the organopolysiloxane molecules to form covalent bonds among the organopolysiloxane molecules. This results in the curing of the silicone rubber composition.

TC-80A (a product name, manufactured by Shin-Etsu Chemical Co., Ltd., thickness: 0.8 mm) is a commercially available heat conductive silicone rubber sheet which is produced by heating and curing a peroxide curing silicone rubber composition. This composition was used in Comparative Experiment 1. The composition was heated and cured by press-molding to produce a peroxide-cured heat conductive silicone rubber sheet TC-A with a thickness of 2 mm.

#### Comparative Experiment 2 (condensation-cured silicone rubber sheet)

A condensation curing silicone rubber composition includes organopolysiloxane molecules with hydrolyzable groups such as silanol groups, alkoxy groups, acyloxy groups, ketoxime groups, and amino groups. When a condensation curing silicone rubber composition is exposed to the air, moisture in the air reacts with the hydrolyzable groups within the organopolysiloxane molecules. The hydrolyzable groups are hydrolyzed to form covalent bonds among the organopolysiloxane molecules. This results in the curing of the silicone rubber composition. Although a condensation curing silicone rubber composition is cured at both ambient temperature and elevated temperature, ambient temperature is typically used. "Air-dry type of curing" in Fick (col. 6, line 59) may be condensation

-4-

curing.

A heat conductive condensation curable silicone RTV rubber composition KE-3467 (a product name, manufactured by Shin-Etsu Chemical Co., Ltd.) was coated and molded on a Teflon sheet. The coated composition was then cured by leaving the composition to stand at room temperature for a week. The cured product was peeled from the Teflon sheet to produce a condensation-cured heat conductive silicone rubber sheet KE-3467' with a thickness of 2 mm.

#### Comparative Experiment 3 (addition-cured silicone rubber sheet)

An addition curing silicone rubber composition includes organopolysiloxane molecules with aliphatic unsaturated hydrocarbon groups (e.g., vinyl groups) bonded to silicon atoms, organohydrogenpolysiloxane molecules with hydrogen atoms bonded to silicon atoms, and a platinum group metal-based catalyst. The platinum group metal-based catalyst accelerates an addition reaction between the silicon atom-bonded aliphatic unsaturated hydrocarbon groups within the organopolysiloxane molecules and the silicon atom-bonded hydrogen atoms within the organohydrogenpolysiloxane molecules to form covalent bonds between the organopolysiloxane molecules and the organohydrogenpolysiloxane molecules. This results in the curing of the silicone rubber composition. Although an addition curing silicone rubber composition is cured at both ambient temperature and elevated temperature, elevated temperature is typically used because the curing is accomplished more quickly.

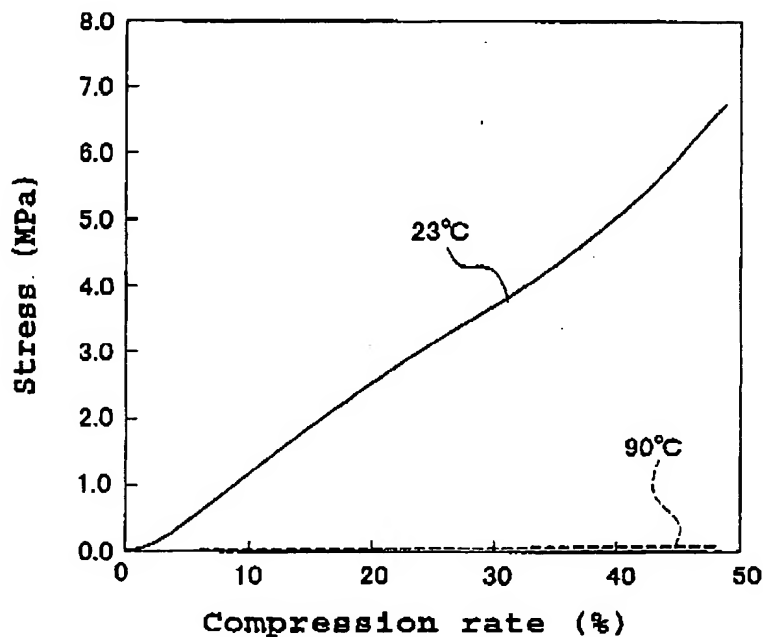
A silicone rubber sheet TC-200TXS (a product name, manufactured by Shin-Etsu Chemical Co., Ltd., thickness: 2 mm) was used as an addition-cured heat conductive silicone rubber sheet. TC-200TXS is a commercially available low-hardness heat-dissipating silicone rubber sheet which is produced by addition-curing.

#### Measurement

-5-

Each of these heat conductive sheets was cut into pieces with a size of 10 mm X 10 mm. One of the pieces was compressed to about 50% of its initial thickness at a rate of 0.5 mm/min with a universal testing machine Autograph AG-I (a product name, manufactured by Shimadzu Corporation). Meanwhile, the stress generated in the piece was measured at each compression rate. The measurements were carried out at temperatures of 23°C and 90°C. The results obtained are given in Figs. 1-4.

Fig. 1: Heat softening, heat conductive sheet K-2' (Experiment 1)



-6-

Fig. 2: Peroxide-cured silicone rubber sheet TC-A (Comparative Experiment 1)

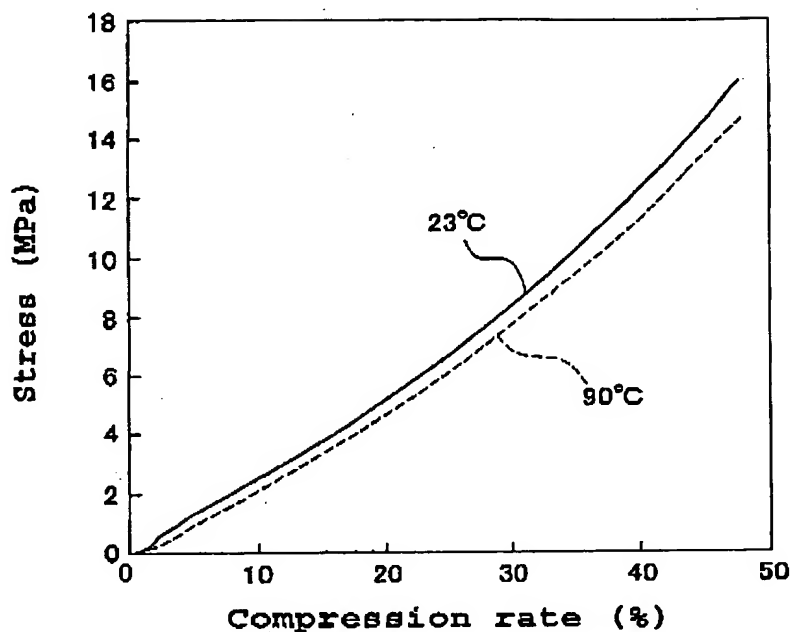
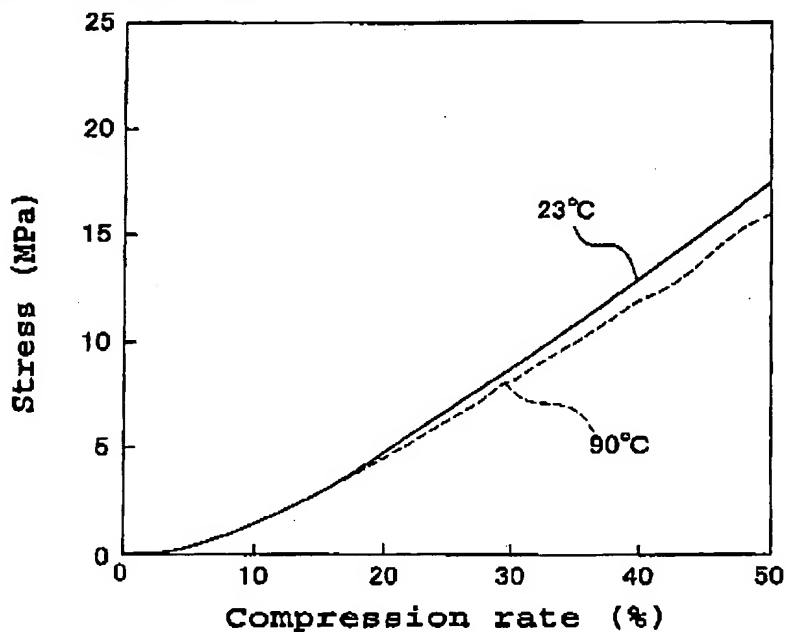
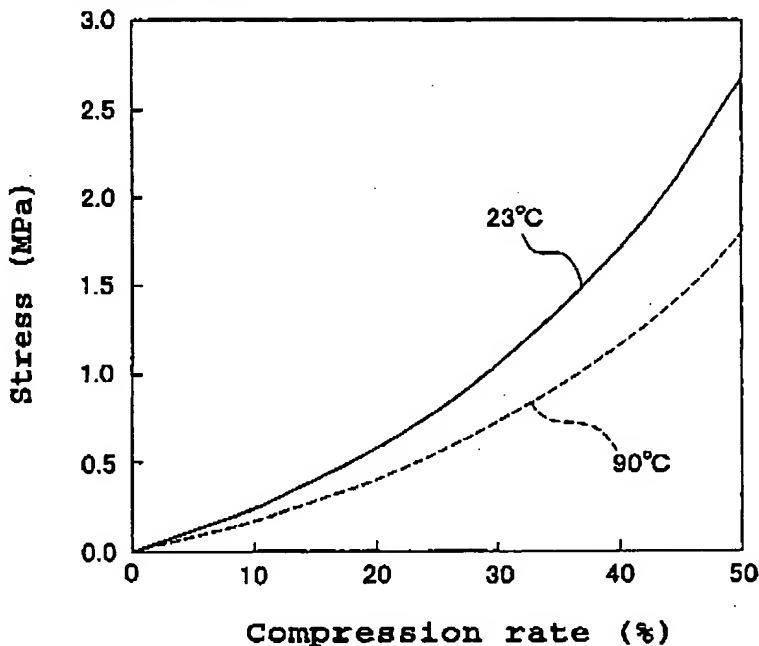


Fig. 3: Condensation-cured silicone rubber sheet KE-3467' (Comparative Experiment 2)



-7-

Fig. 4: Addition-cured silicone rubber sheet TC-200TXS  
(Comparative Experiment 3)



#### 7. Conclusion

Fig. 1 shows that, at 23°C, the stress becomes larger as the compression rate increases. In contrast, at 90°C, the stress is almost 0 MPa even if the compression rate increases to about 50%. These results indicate that the heat softening, heat conductive sheet K-2' is essentially solid at room temperature but melts at 90°C. In other words, the sheet cannot be cured or crosslinked at 90°C.

Figs. 2-4 show that, at both temperatures 23°C and 90°C, the stress becomes larger as the compression rate increases. The stress is reduced by only about 10% (Comparative Experiments 1 and 2) or by only about 30% (Comparative Experiment 3) at the same compression rate when the temperature rises from 23°C to 90°C. These results indicate that the peroxide-cured silicone rubber sheet TC-A; the condensation-cured silicone rubber sheet KE-3467', and the addition-cured

-8-

silicone rubber sheet TC-200TXS become slightly softer but do not melt when the temperature rises from 23°C to 90°C.

As can be seen from the above, pre-cured silicone rubber sheets generally do not melt at a temperature that ranges from 40°C to 100°C. Therefore, the pre-cured upper layer 7A of Fick is not capable of melting at a temperature that ranges from 40°C to 100°C. Thus, the melting behavior of the layer (a) of the present application is unexpected from Fick.

8. I, the undersigned petitioner, declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001, of title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

Date: July 18, 2006

Ikuo Sakurai

Ikuo SAKURAI